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G. Mankey – University of Alabama

et al.

Citation of published version:

Mankey, G., et al. (1991): The Effect of Microstructure on the Magnetic Behavior of Epitaxial Cobalt Layers. *Journal of Vacuum Science and Technology A,* 9(3).
DOI: https://doi.org/10.1116/1.577666

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Citation: Journal of Vacuum Science & Technology A **9**, 1595 (1991); doi: 10.1116/1.577666
View online: https://doi.org/10.1116/1.577666
View Table of Contents: https://avs.scitation.org/toc/jva/9/3
Published by the American Vacuum Society

The effect of microstructure on the magnetic behavior of epitaxial cobalt layers

G. J. Mankey, M. T. Kief, and R. F. Willis
Physics Department, The Pennsylvania State University, University Park, Pennsylvania 16802

(Received 17 September 1990; accepted 29 October 1990)

We have studied the magnetic behavior of thin cobalt films epitaxed on a Cu (001) substrate as a function of their growth temperature. At 150 K, the film texture is rough and the surface coverage is incomplete for film thicknesses < 1.2 monolayers. Smoother films are obtained as the substrate temperature is increased, but at the expense of increased copper interdiffusion. Growth of films at 450 K produces smooth continuous epitaxial layers but coated with a layer of interdiffused copper which serves to lower the surface free energy. Copper interdiffusion can be controlled at intermediate temperatures between 300 and 450 K. We report on the effect of annealing cycles on the microstructure of these films in relation to their magnetic properties, as revealed by surface magneto-optic Kerr effect hysteresis loop behavior.

I. INTRODUCTION

There has been much recent interest in the magnetic behavior of thin ferromagnetic films epitaxed on nonmagnetic surfaces.[1] The subject is a controversial one, however, with various groups reporting different behavior due to the difficulty of preparing films with sufficient purity and microstructural perfection. These difficulties are compounded in systems in which the magnetic moments are sensitive to film strain [Fe/Cu (001)][2] or have a tendency to alloy with the substrate [Ni/Cu (001)].[3] We have chosen to study in some detail cobalt layers epitaxed on Cu (001) crystals since the cobalt moment is known to be stable to film strain[4] and cobalt does not form a stable bulk alloy phase.[3] We have characterized the morphology of films over a thickness range extending from fractions of a monolayer (ML) up to many MLs using a variety of methods.

In this paper, we report measurements of variations in the magnetic properties as a function of film growth temperature and film thickness. Changes in film morphology were monitored by hydrogen gas titration studies, low-energy electron diffraction (LEED) and reflection high-energy electron diffraction (RHEED) oscillations, Auger and ultraviolet photoelectron spectroscopy. Magnetic properties, including film magnetization M, and coercivity H_c, and Curie Temperature T_c, were obtained with a surface magneto-optic kerr effect (SMOKE) instrument.[5] All measurements were performed *in situ* in a single ultrahigh vacuum (UHV) system with a base pressure of 5×10^{-11} mbar.

II. EXPERIMENT

A schematic of the experimental system is shown in Fig. 1. The Cu (001) single crystal substrate is supported on a commercial Vacuum Generators HPLT600/SM2 sample manipulator with facilities for heating to 1000 K and cooling to 150 K. Successive cycles of 500 eV Ar ion bombardment, followed by anneals to 1000 K, produced a sharp $p(1 \times 1)$ LEED pattern. Auger spectroscopy using an Omicron single-pass cylindrical mirror analyser (CMA) was used to monitor contamination. The cobalt evaporation sources were housed in a separate UHV chamber with a water cooled shroud so that they can be extensively degassed prior to use. The evaporation sources were emission current stabilized to produce a constant flux at the sample equivalent to 1 ML of material deposited in 3 min. The evaporation flux was monitored with a quartz crystal microbalance (QCM) located at a quarter of the source/substrate separation for enhanced sensitivity. Precise determination of the film thickness was achieved by comparing simultaneous QCM thicknesses and RHEED oscillations with Auger spectra. The RHEED oscillations were obtained using the 5 keV electron gun coaxial within the CMA at an incident angle of 82° with respect to the surface normal. These measurements were found to be consistent with film coverage and roughness effects observed using hydrogen titration results. Analysis of hydrogen desorption spectra reveal changes in the films microstructure. The deposited films could be rapidly moved to other positions in the apparatus (Fig. 1) for further analysis using SMOKE, LEED, and photoelectron spectroscopy.

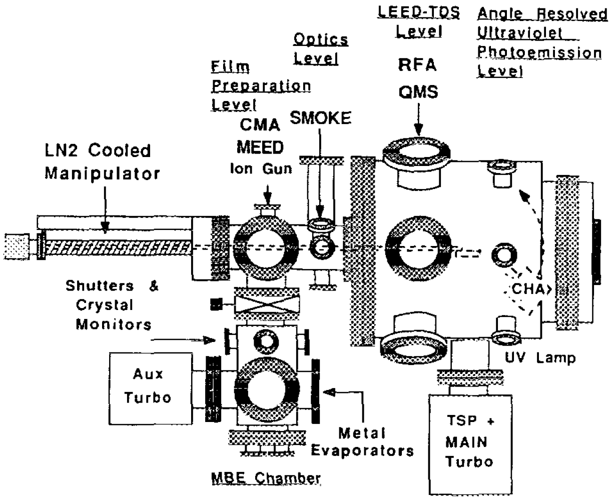

FIG. 1. Schematic diagram of experimental apparatus, showing various interconnected ultrahigh vacuum chambers for ultrathin magnetic film growth and characterization.

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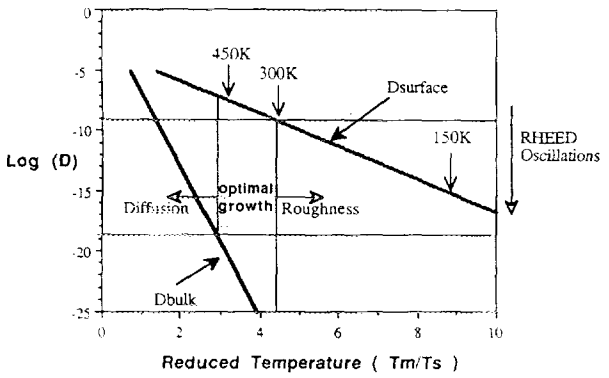

FIG. 2. Diagram illustrating the dependance of surface and bulk diffusion on reduced temperature, T_m/T_s, for metals (T_m = melting temperature; T_s = substrate temperature). Adapted from Ref. 6.

III. RESULTS AND DISCUSSION

Figure 2 shows the dependence of surface and bulk diffusion on reduced temperature T_m/T_s for metals[6] (T_m = melting temperature; T_s = substrate growth temperature). The shaded region defines the optimal growth conditions for thin film overlayers, which is dependent upon evaporation rate. Films grown at higher temperatures than the shaded region suffer from interdiffusion from the bulk. Films grown at too low a temperature are rough because surface diffusion is too slow. Between about 300 and 450 K there is a narrow window for optimal growth. In this region films grow layer-by-layer with an abrupt interface and with well-defined RHEED oscillations. We therefore chose to study the properties of films deposited at 150, 300, and 450 K.

LEED and RHEED patterns confirm that the 150 K deposited layers are rough on a microscopic scale.[7] Films grown at 300 K produce much sharper $p(1\times1)$ diffraction patterns and a lower background intensity. Films prepared at 450 K substrate temperature have a sharp $p(1\times1)$ pattern indistinguishable from that of the well-prepared Cu(001) single crystal substrate. Typical RHEED oscillations as a function of film deposition at 150, 300, and 450 K are shown in Fig. 3. The RHEED oscillations observed for the 450 K (and 300 K) films are indicative of layer-by-layer growth,[8] particularly for film thicknesses above 2 ML. These oscillations are less discernible for films deposited at temperatures below 300 K.

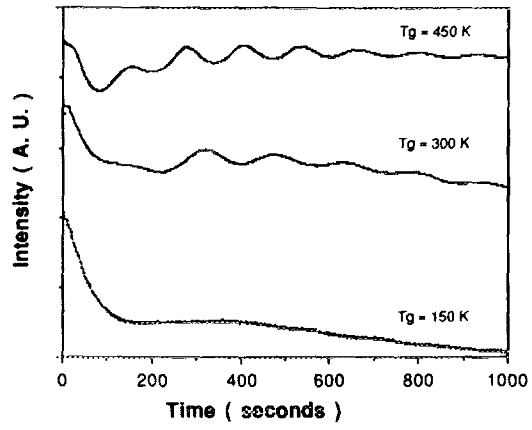

FIG. 3. Comparison of RHEED oscillations at three substrate growth temperatures. The diffraction conditions were identical for each measurement with an incident electron energy of 5 keV, incident angle of 82° along the (01) azimuth.

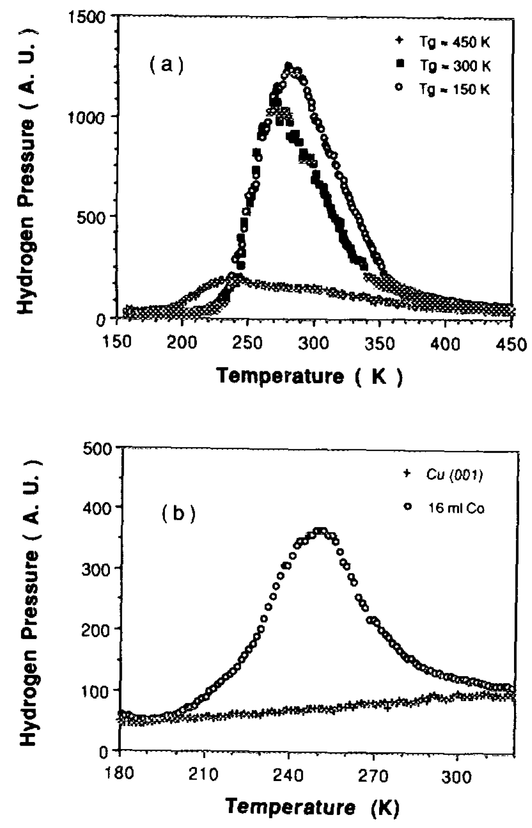

FIG. 4. (a) Hydrogen thermal desorption spectra for 2 ML Co films grown at different substrate temperatures: T_g = growth temperature. (b) Thermal desorption spectra of hydrogen from Cu (001) and bulk (16 ML) fcc cobalt.

Typical hydrogen thermal desorption spectra (TDS), after dosing 2 ML thick films to saturation coverage, are shown for T_s = 150, 300, and 450 K in Fig. 4(a), and compared to those for clean Cu (001) and a thick (16 ML) "bulk" fcc cobalt film in Fig. 4(b). As can be seen from Fig. 4(b), hydrogen gas titration is a convenient method for studying the morphology of these films since the sticking coefficients for hydrogen chemisorption is much lower on Cu (001) than on cobalt.[9] Hydrogen coverage-dependent TDS studies confirm that the desorption kinetics are second order.[10] In sharp contrast to the 150 and 300 K films, the film grown at 450 K shows no hydrogen desorption peak but only a broad background [Fig. 4(a)]. In fact, the 450 K spectrum shown in Fig. 4(a) is more indicative of thermal desorption from the copper [Fig. 4(b)]. The film deposited at 450 K is very close to the upper optimal limit for which bulk diffusion begins to become important (Fig. 2). The absence of a hydrogen desorption peak at 280 K for 450 K deposited films suggests that copper has segregated on to the surface of the cobalt thin film to lower the surface free energy.[11] This is confirmed by angle-resolved ultraviolet photoelectron spectra (Fig. 5). Since the clean Cu (001) surface has a well-characterized Tamm surface state spectral feature

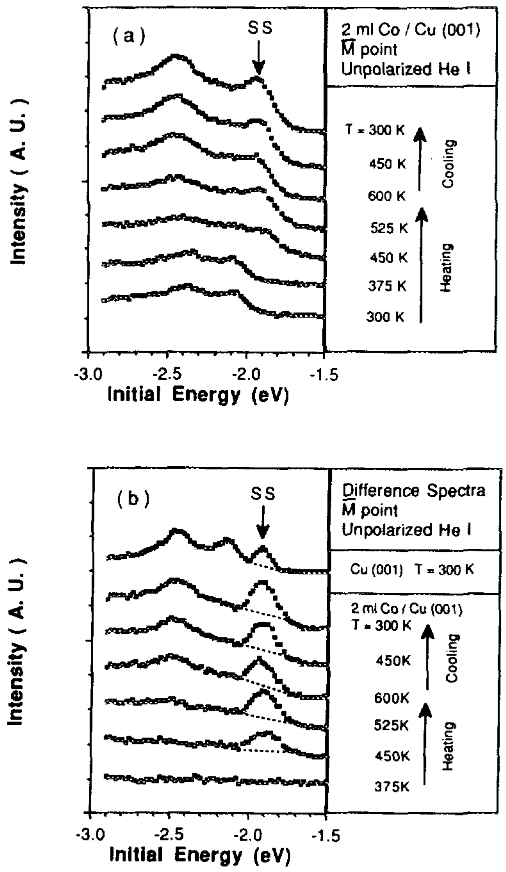

FIG. 5. (a) Photoemission energy distribution curves at the $\bar{M}$ point of the Brillouin zone for a 2 ML Co/Cu (001) film showing the effect of annealing to 600 K and cooling back to 300 K. (b) Difference curves obtained by subtracting the energy distribution curve of the as-grown film from subsequent curves

1.9 eV below the Fermi energy located at Mbar in the surface Brillouin zone[12] and cobalt has no such feature, the appearance of a similar peak, *SS*, in the spectrum of the 2 ML thick cobalt film at this angle and energy confirms the presence of a copper overlayer. There is no noticeable difference observed in the spectra of the as-grown films at 300 K and after annealing to 400 K. However, there is a significant irreversible change in films annealed at 450 K. Similarly, films grown at 450 K also show the copper surface state spectral feature. RHEED oscillations are observed for all films grown between 300 and 450 K; in the latter case, the RHEED oscillations do not distinguish the copper surface segregation.

These differences in film microstructure are reflected in their magnetic properties. Magnetization versus magnetic field hysteresis behavior was monitored using SMOKE. Both in-plane and perpendicular magnetizations were investigated using the longitudinal and polar Kerr effects. The magnetic field was typically varied up to 125 G at frequency of 0.1 Hz. A typical hysteresis curve obtained from a 2 ML film of Co/Cu (001) grown at a substrate temperature on 300 K is shown in Fig. 6.

All of the Co/Cu (001) films show in-plane magnetization for all thicknesses. The magnetization M and, in particular, the Curie temperature are a sensitive function of film thickness, as shown in Fig. 7. The films prepared at 300 K show a linear rise in T_c with thickness. Films prepared at 450 K show a shallower slope. For thicknesses above ~ 1.25 ML the 450 K films have a consistently lower T_c than do the 300 K films. We note that the effect of the copper overlayer is to lower the magnetization of the 450 K grown cobalt films.[13] These results are in substantial agreement with similar results for films deposited at 450 K reported by Schneider *et al.*[14] Similarly, hydrogen adsorption changes the magnetization, reducing M and T_c.[15]

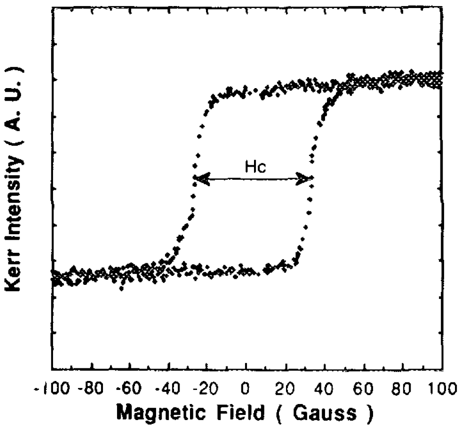

FIG. 6. A typical hysteresis curve for a 2 ML cobalt film.

The coercive field H_c vs T behavior of a 2 ML film deposited at 150, 300, and 450 K is shown in Fig. 8. Again, we see that the largest discrepancy in behavior is observed for the film grown at $T = 450$ K; H_c remains independent of T and has the low value of $H_c \approx 15$ G. In contrast, both the "rough" 150 K and the "smoother" 300 K films start with a much higher value of $H_c \approx 30$ G, which anneals irreversibly down to the 15 G, 450 K value. Cooling the as-grown 300 K film to 150 K and then warming to 450 K follows the general behavior of the 150 K deposited film. This indicates that micromagnetic structure of the 150 and 300 K films are similar, despite the fact that the 150 K appears to be more microscopically rough.[16] The irreversible change brought about by annealing or growth at 450 K is due to copper surface segregation.

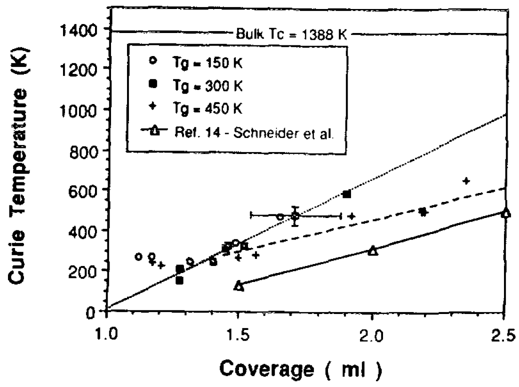

FIG. 7. Dependance of the Curie temperature on film thickness and growth temperature.

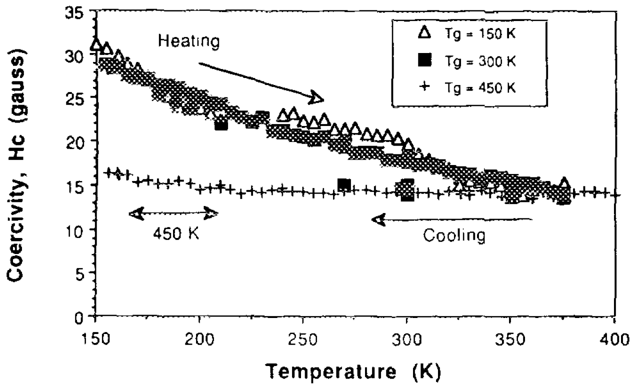

FIG. 8. Variation of coercive field with temperature for 2 ML Co/Cu (001) films grown at different substrate temperatures.

IV. CONCLUDING REMARKS

These results show that metastable magnetic thin films of uniform thickness can be grown on nonmagnetic metallic substrates providing due care is taken to establish the growth conditions. The magnetic properties are sensitive to the film morphology. Interface diffusion can be limited by careful choice of film growth parameters, such that uniform layers can be obtained down to film thicknesses of a few monolayers. Disparities between the results reported by different groups reflect differences in the growth conditions and/or film thickness calibration.

ACKNOWLEDGMENT

This work was supported by a grant from the National Science Foundation No. NSF-DMR881884.

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